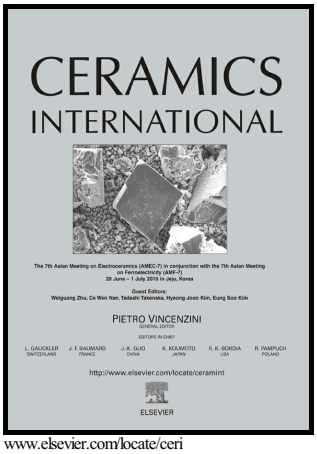

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Synthesis of single-phase $Ca_3Co_4O_9$ ceramics and their processing for a microstructure-enhanced thermoelectric performance

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Abstract

Single-phase $Ca_3Co_4O_9$ with a high porosity, having a ZT of 0.09 at 627°C, was successfully prepared by a simple solid-state reaction using fine powders of $CaCO_3$ and Co_3O_4 after a calcination at 760 °C for 12 hours. It was excellent either for the preparation of the starting powder for the processing of the $Ca_3Co_4O_9$ ceramics or for direct processing. The influence of already-reported processing methods (classic sintering, hot pressing, free-edge spark-plasma sintering (SPS) of a sintered pellet) and new methods, such as free-edge SPS of a pellet from just-compacted powder, cold pressing of a sintered pellet without post annealing, and free-edge cold pressing of a sintered pellet with post-annealing, on the preparation of $Ca_3Co_4O_9$ ceramics was studied. The results showed how the density, the grain morphology and the microstructural anisotropy can all influence the thermoelectric characteristics of $Ca_3Co_4O_9$ ceramics measured in directions parallel and perpendicular to the applied pressure. While the fully dense (99%) and perfectly textured ceramics prepared by the free-edge SPS of a pellet from just-compacted powder had a ZT of 0.17, the highest ZT of 0.31 was obtained for the free-edge cold-pressed and annealed ceramics with modest texturing and low density (65%), having a very low κ of 0.47 W/mK. The results also showed that thin and irregular-shaped plate-like grains with sharp edges are preferred, while their thickening accompanied by rounding of their shape resulted in a reduced thermoelectric performance. The study revealed both the possibilities and the limitations for enhancing the TE characteristics of $Ca_3Co_4O_9$ ceramics just through microstructure optimisation.